UCRL-ID-143346

Plutonium Immobilization Task 5.6 Metal Conversion: Milestone Report - Perform Feasibility Demonstrations on Pu-Al Alloys

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April 12, 2001

U.S. Department of Energy

Lawrence Livermore National Laboratory

DISCLAIMER

This work was performed under the auspices of the U. S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

Date: 28 December 2000

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Introduction

The Plutonium Conversion Task within the Plutonium Immobilization Program (PIP) transforms incoming plutonium (Pu) feed materials into an oxide acceptable for blending with ceramic precursors. One of the feed materials originally planned for PIP was unirradiated fuel, which consisted mainly of the Zero Power Plutonium Reactor (ZPPR) fuel. Approximately 3.5 metric tons of Pu is in ZPPR fuel. The ZPPR fuel is currently stored at the Argonne National Laboratory - West as stainless steel clad metal plates and oxide pellets, with the vast majority of the Pu in the metal plates. The metal plates consist of a Pu-U-Mo alloy (containing 90% of the ZPPR plutonium metal) and a Pu-Al alloy (containing 10% of the ZPPR plutonium metal). The Department of Energy (DOE) decided that ZPPR fuel is a national asset and, therefore, not subject to disposition. This report documents work done prior to that decision.

The Hydride-Oxidation (HYDOX) Process was selected as the method for Metal Conversion in PIP because it provides a universal means for preparing oxide from all feed materials. HYDOX incorporates both the hydride process, originally developed to separate Pu from other pit materials, as well as the oxide formation step. Plutonium hydride is very reactive and is readily converted to either the nitride or the oxide. A previous feasibility study demonstrated that the Pu-U-Mo alloy could be successfully converted to oxide via the HYDOX Process. Another Metal Conversion milestone was to demonstrate the feasibility of the HYDOX Process for converting plutonium-aluminum (Pu-Al) alloy in ZPPR fuel plates to an acceptable oxide. This report documents the results of the latter feasibility study which was performed before the DOE decision to retain ZPPR fuel rather than immobilize it.

ZPPR Fuel Plate Processing Objectives

Three objectives were defined for the processing of ZPPR fuel plates to demonstrate the feasibility of the process for this type of material:

1. Assure the material will hydride and convert to oxide
2. Assure the removal of Pu from the cladding
3. Assure that cladding remains in the basket

The results reported here are measured against these criteria in assessing the successfulness of the processing experiment conducted.

Experimental

The HYDOX reactor vessel used in all of the experiments is shown in Figure 1. The vessel is composed of a reaction chamber containing a basket which can be rotated during the process of converting the alloy or metal to the oxide. The oxide material falls by gravity through the basket mesh and collects in a heated crucible. The crucible contains a quartz or Hastelloy frit that allows reaction gas to pass through it.

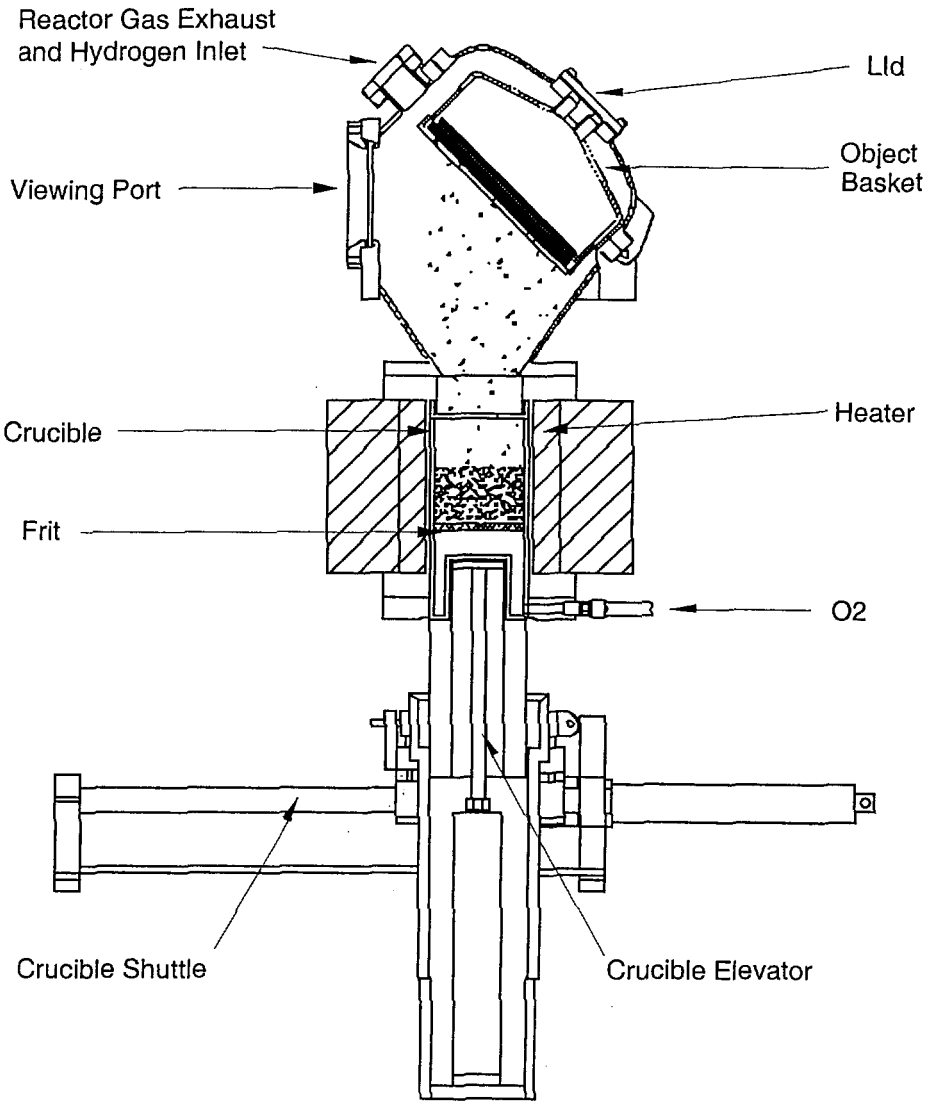

Figure 1. Schematic depiction of the HYDOX reactor vessel components

Two ZPPR fuel processing experiments were conducted using Pu-Al alloy plates and a three-step HYDOX process: metal hydride formation was followed by conversion to metal nitride and, finally, conversion to metal oxide. Tables 1-3 summarize the ZPPR plate characteristics, process parameters, and results.

Table 1. ZPPR plate characteristics

plate #	dimensions (inch.)	plate type	standard core (g)	total Pu (g)	total Al (g)	total Am (g)
1	1/8 x 2 x 3	Pu-Al	105.3	202.0	2.3	0.5
2	1/8 x 2 x 3	Pu-Al (Ni plated)	105.2	197.9	2.4	0.5

Table 2. Processing parameters

plate #	run date	plates /run	net feed /run (g)	SS shell /run (g)	pieces cut	hydr. time (hr)	nitrid. time (hr)	oxid. time (hr)	oxid. temp (°C)	oxid. flow direction
1	1/00	2	204.7	47.4	6	0.5	0.4	0.5	700	O_2-He down
2	2/00	2	200.7	57.8	8	0.75	0.6	1.0	600	O_2-He down

Table 3. Processing results

plate #	PuO_2 (g) theory	Al_2O_3(g) theory	AmO_2(g) theory	Total Oxide (g) theory	Actual Oxides (g)	cladding empty	oxide appear.
1	229.0	4.3	0.5	233.8	233.3	No	green
2	224.4	4.5	0.5	229.4	228.4	Yes*	green

* Requires tumbling or manual manipulation

Pu-Al alloy Plates 1 and 2 were thinner (0.125") than standard ZPPR plates (0.25"). The plates were cut with a hacksaw as shown in Figures 2 and 3, respectively. The metal hydride was subsequently converted to nitride and then to oxide in the crucible. The oxygen-helium flow rate during the oxidation step was 1.0 L/min O_2 and 10 L/min He. Due to the presence of a heater designed for use in experiments involving weapon parts, the basket was held stationary rather than rotated.

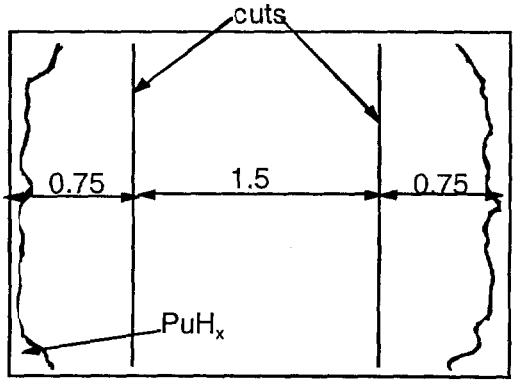

Figure 2. Plate 1 cutting pattern and location of residual PuH_x

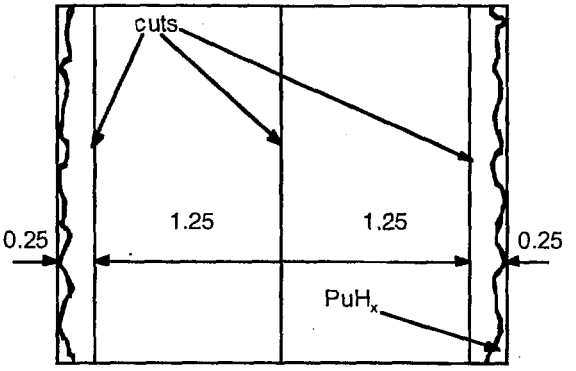

Figure 3. Plate 2 cutting pattern and location of residual PuH_x

Results/Discussion

In the course of processing Plate 1, Objective 2, removal of all Pu from the cladding, was not achieved. The 0.75" end cuts on the plate retained plutonium hydride (Figure 2). It is likely that the thinness of the plates in combination with the depth of the pockets caused the plutonium hydride to be retained. The cladding in the middle section of the plate buckled (see Figure 4) during cutting, thus exposing the entire Pu surface in this area to hydrogen and leading to an accelerated hydration rate of 1.5 hrs/kg.

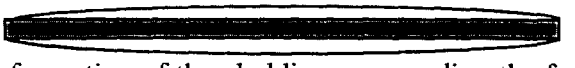

Figure 4. Deformation of the cladding surrounding the fuel in Plate 1

The pocket depths at the edges of Plate 2 were reduced from 0.75" to a shallower 0.25" depth in an effort to eliminate the retention of PuH_x in the end pieces. While some PuH_x was still trapped inside the nickel plating (Figure 3), manual manipulation of these pieces released the trapped hydride. In the prototypical system, the part heater does not preclude the rotation of the basket needed to agitate material and release the plutonium hydride from inside the cladding. It is believed that the nickel plating is sturdy enough to withstand basket rotation, and hence metal-hydride could be removed from the pockets by tumbling the clad pieces in the basket at the completion of the hydride formation step, thus satisfying Objective 2. Additionally, the only limit to a further reduction in the strip width is the dimension of the basket openings, to assure the cladding is retained in the basket during rotation. The basket holes were originally selected to retain 0.25" strips, but a smaller hole dimension could help further insure the retention of the clad pieces during the processing of smaller strips.

In both experiments, nearly 100% of the oxide was recovered, demonstrating clearly the feasibility of the HYDOX Process for conversion of the Pu-Al fuel.

Conclusions

The first and third objectives of this feasibility demonstration were meet with both demonstration tests. The Pu-Al alloy hydrided and converted to oxide similar to low alloyed plutonium metal and the cladding material stayed behind in the basket. The second objective of removing all the plutonium from the cladding was not achieved in the first test from the 3/4 inch end sections. These end sections were not prototypical, therefore, in the second test prototypical end sections of 1/4 inch were used. In the second test only a little material remained in the 1/4 inch end sections which was readily released when lightly tapped. In a tumbled basket, such as in the prototypical HYDOX system, hydride or nitride will definitely be released from the cladding ends due to the repeated impacts of the cladding against the basket. Therefore, the feasibility demonstration showed that the HYDOX process would remove the material from sheared Pu-AL ZPPR plates, separate the plutonium from the cladding material and convert the plutonium to an oxide.